Theory for the Transient Statistics of a Dye Laser

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A theory is presented to explain the statistical properties of the growth of dye-laser radiation. Results are in agreement with recent experimental findings. The different roles of pump-noise intensity and correlation time are elucidated.

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In recent years the dye laser has been the subject of great interest, becoming a prototype nonequilibrium system in which random fluctuations of a control parameter are known to be important. Indeed, anomalous steady-state statistical properties of dye lasers have been explained in terms of stochastic models which include a fluctuating pump parameter.¹ Recent experiments² on the transient statistics of dyelaser radiation evidence two separate time regimes dominated respectively by quantum and pump noise. The first regime is associated with the behavior close to the initial unstable state and the second with the behavior close to steady state. Evidence for two time scales is also obtained through the study of the timedependent properties of the dye-laser intensity.³ In the models used to study steady-state properties, quantum noise has been generally neglected, although it has been shown that it might be important in some cases (Lett, Short, and Mandel¹). Quantum noise is, however, essential to describe the transient behavior associated with the decay of an unstable state. In fact, the experiments by Roy, Yu, and Zhum^{2, 3} confirm our earlier suggestion,⁴ based on the analysis of a simple bistable one-variable model with multiplicative white noise, that transient experiments could be used to disentangle the effect of multiplicative and additive noise.

In this Letter we present a theory for the transient radiation statistics of the single-mode dye laser. Our theory considers the stochastic process of the complex field amplitude $E = E_1 + iE_2$, and it is based on a simple direct approximation for this process. Within this theory we obtain results which are relevant for the two approaches generally used to describe transient radiation statistics. The first approach focuses on the time dependence of statistical properties of the laser intensity, and in particular on the transient enhancement of its variance.⁵ The second approach, followed by Roy, Yu, and Zhum,² characterizes the relaxation process in

terms of the statistical properties of the times for which the laser intensity has grown to a given value (passage times).⁶ An earlier analysis of passage-time statistics is due to Fox.⁷ Our results for the passagetime statistics are in good quantitative agreement with the experimental results. The variance of the passage-time distribution is seen to be sensitive to pump noise even close to the unstable state. In addition, we describe the role played by the two parameters which determine the pump noise (noise intensity Dand correlation time τ). We find that the effect of pump noise is partially compensated as a result of its finite correlation time. As regards statistical properties of the laser intensity we predict a reduction of anomalous fluctuations due to pump noise. This reduction is again partially compensated by the finite correlation time τ . We finally propose experiments closer to threshold than those of Ref. 2, to get a stronger evidence of all these effects.

Our dynamical model is given by the following Langevin equation for the complex field amplitude E:

$$\partial E(t)/\partial t = a(t)E - A|E|^2E + \sqrt{\epsilon}\xi(t).$$
(1)

Here $\xi(t)$ is a complex Gaussian white noise with temporal correlation $\langle \xi(t)\xi^*(t')\rangle = 2\delta(t-t')$ which accounts for quantum noise. Pump fluctuations are taken into account in the parameter $a(t) = a_0 + \sqrt{D} \eta(t)$ where $\eta(t)$ is a complex Gaussian noise with zero mean and correlation

$$\langle \eta(t)\eta^*(t')\rangle = (1/\tau)\exp[-|t-t'|/\tau].$$

The validity of this model to describe the transient dynamics of the dye laser can be considered as well established after the good comparison obtained between experimental results and numerical simulations by Roy, Yu, and Zhum.² The relevant physical quantity is the laser intensity $I(t) = |E|^2$. A solution of (1) for the process I(t) can be obtained when quantum noise is neglected:

$$I(t) = I(0) \exp\{2[a_0 t + \omega(t)]\} / \left[1 + 2AI(0) \int_0^t dt' \exp\{2[a_0 t' + \omega(t')]\}\right]$$
(2)

where $\omega(t) = \sqrt{D} \int_0^t dt' \operatorname{Re}\eta(t')$. The solution can be seen as a mapping between the value of I(t) at time t and the value at the initial time I(0) for a given realization of the pump noise $\eta(t)$. Quantum noise neglected in (2) is

essential to trigger the decay from the unstable state I(0) = 0. Close to this state nonlinear terms and pump fluctuations can be neglected in (1). The solution of (1) in such a regime can be written as

$$E(t) = e^{at}h(t), \quad h(t) = \sqrt{\epsilon} \int_0^t dt' e^{-at'}\xi(t'). \quad (3)$$

The complex process h(t) is a bivariate Gaussian process of zero mean and variance $\langle |h|^2 \rangle = (\epsilon/a_0) \times (1 - e^{-2a_0t})$. For times much larger than a_0^{-1} , h(t)becomes a time-dependent random variable h with variance ϵ/a_0 . In this time regime the realizations of the stochastic process E(t) are very close to trajectories in which quantum noise vanishes but with a random initial condition E(0) = h. Our approximation for the process defined by (1) is then to use the mapping (2) but with the initial condition replaced by the process h(t). Explicitly we have

$$I(t) = \frac{|h|^2(t)\exp\{2[a_0t + \omega(t)]\}}{1 + 2A|h|^2(t)\int_0^t dt'\exp\{2[a_0t' + \omega(t')]\}}.$$
(4)

This gives rise in a natural way to a process which is dominated by quantum noise close to the unstable state (early times) and by pump noise after leaving this state. Our approximation is a generalization of the quasideterministic theory.⁸ This theory has been successfully applied to the study of the transient statistics of an ordinary laser intensity.⁹ An extensive analysis of the validity of this extension for a simple process involving multiplicative noise was given in Ref. 4.

We first obtain the statistical properties of passage times from our approximation given by (4). One is in general interested in passage times which determine the time regime in which the system leaves the unstable state. This regime corresponds to an intensity range beyond the immediate vicinity of the unstable state and also far from the final equilibrium state. The first condition allows us to approximate h(t) by h.

$$W(\lambda) = W_{D=0}(\lambda) \exp\left\{\lambda^2 T_0 \frac{D/[2a_0^2(1+\lambda D/a_0^2)]}{(1+\lambda D/a_0^2)^{1/2}}\right\}$$

From this it follows that

$$\langle t \rangle = \langle t \rangle_{D=0} + D/2a_0^2 = T_0 - (1/2a_0)\psi(1) + D/2a_0^2,$$
(9)

$$\langle (\Delta t)^2 \rangle = \langle t^2 \rangle - \langle t \rangle^2 = \langle (\Delta t)^2 \rangle_{D=0} + \frac{DT_0}{a_0^2} + \frac{D^2}{2a_0^4} = \frac{1}{4a_0^2} \psi'(1) + \frac{DT_0}{a_0^2} + \frac{D^2}{2a_0^4}, \tag{10}$$

where $\psi(1)$ is the digamma function.¹²

In the situation of actual interest in which the pump noise is not white $(\tau \neq 0)$, (6) cannot be explicitly solved for t. However, in the normal situation in which τ is small compared with typical passage times, D(t) can be approximated as $\sigma(\tau) \sim \sigma(t-\tau)$ and we can proceed as above. In the opposite limit $\tau >> t$, D(t) = 0 so that the effect of pump noise disappears. The second condition allows us to neglect nonlinear terms in our mapping. Equation (4) is then approximated by¹⁰

$$I(t) \sim |h|^2 \exp\{2[a_0 t + \omega(t)]\}.$$
 (5)

In order to obtain the passage time t to a given reference value of I we note that the values taken by the process $\omega(t)$ at time t can be obtained replacing $\omega(t)$ by $\nu \sqrt{\sigma(t)}$, where ν is a Gaussian variable of zero mean and unit variance, and $\sigma(t) = \langle \omega^2(t) \rangle = Dt$ $-D\tau(1-l^{-t/\tau})$. Equation (5) then gives

$$t + \nu \sqrt{\sigma(t)} = \ln(I/|h|^2).$$
 (6)

Solving (6) for t gives the passage times as a function of the stochastic variables h and ν . Statistical properties can be obtained from the generating function $W(\lambda) = \overline{W}_{\nu}(\lambda) = \langle \overline{e^{-\lambda t}} \rangle$, where the bar indicates average over ν and the angular brackets average over the distribution of h. For the sake of clarity we first consider the white-noise limit $(\tau = 0)$ for $\eta(t)$. In this limit (6) is easily solved for t. At this point we recall that in the absence of pump noise (D=0) the asymptotic value for small ϵ of the mean first-passage time is $T_0 = (1/2a_0)\ln(a_0I/\epsilon)$. This is a large quantity for small ϵ . Expanding the solution of (6) for t to second order in $D^{1/2}\nu/2a_0T_0^{1/2}$ and to first order in $\ln(|h|^2 a_0/\epsilon)/2a_0 T_0$ we obtain after averaging over h

$$W_{\nu}(\lambda) = W_{D=0}(\lambda) \exp\left\{-\lambda \left[\frac{D\nu^2}{2a_0^2} - \frac{D^{1/2}T_0^{1/2}\nu}{a_0}\right]\right\}$$
(7)

 $W_{D=0}(\lambda) = \Gamma[(\lambda/2a_0) + 1]e^{-\lambda T_0}$ is where the first-passage-time generating function in the absence of pump noise. The result for $W_{D=0}(\lambda)$ was also derived by Haake, Haus, and Glauber.¹¹ Averaging over ν without further approximation gives

$$\rangle = \langle t^2 \rangle - \langle t \rangle^2 = \langle (\Delta t)^2 \rangle_{D=0} + \frac{DI_0}{a_0^2} + \frac{D^2}{2a_0^4} = \frac{1}{4a_0^2} \psi'(1) + \frac{DI_0}{a_0^2} + \frac{D^2}{2a_0^4}, \tag{10}$$

For $\tau \ll t$ we obtain the same results (7) and (8) but with T_0 replaced by $T_0 - \tau$ except in the factor $W_{D=0}(\lambda)$. As a consequence $\langle t \rangle$ turns out to be independent of τ in this approximation and

$$\langle (\Delta t)^2 \rangle = \langle (\Delta t)^2 \rangle_{D=0} + \frac{D(\tau_0 - \tau)}{a_0^2} + \frac{D^2}{2a_0^4}.$$
 (11)

The correction due to multiplicative noise for $\langle t \rangle$ is typically very small, so that the mean passage time has a value essentially determined by quantum fluctuations. For the variance we have obtained a negligible contribution $D^2/2a_0^4$ and an important enhancement due to the factor T_0 in the DT_0/a_0^2 contribution. This enhancement is reduced by the colored character of pump noise, in agreement with the fact that the effect of pump noise disappears as $\tau \rightarrow \infty$. Such a reduction becomes larger the larger the noise intensity D. The values of the parameters of model (1) fitted to experiments are² $a_0 = 2.16 \times 10^6$ s⁻¹, $A = 2.64 \times 10^6$ s⁻¹, $\epsilon = 0.0043$ s⁻¹, $D = 3 \times 10^4$ s⁻¹, $\tau^{-1} = 2.4 \times 10^6$ s⁻¹. For these values of the parameters and a reference value of $I_r = 0.3 a_0 / A$ we obtain from (9) and (11) $\langle t \rangle = 4.45 \,\mu s, \quad \langle (\Delta t)^2 \rangle = 11.32 \times 10^{-2} \,\mu s^2, \quad \langle (\Delta t)^2 \rangle$ - $\langle (\Delta t)^2 \rangle_{D=0} = 2.51 \times 10^{-2} \,\mu s^2.$ These numbers agree very well with the results in Ref. 2. We note that the contribution to $\langle (\Delta t)^2 \rangle$ due to the multiplicative noise is around 20% and the negative contribution to $\langle (\Delta t)^2 \rangle$ due to the finite correlation time is only about 2%. These differences cannot be distinguished within the accuracy of the experimental data. We have thus done simulations with larger values of D. According to (9) for a value of D ten times larger $\langle t \rangle$ changes by less than 1%, while $\langle (\Delta t)^2 \rangle$ becomes dominated by pump noise, $\langle (\Delta t)^2 \rangle = 34.06 \times 10^{-2} \ \mu s^2$, $\langle (\Delta t)^2 \rangle - \langle (\Delta t)^2 \rangle_{D=0} = 25.25 \times 10^{-2} \ \mu s^2$; and the negative contribution due to the finite correlation time is close to 10%. Figure 1 reports a comparison of the numerical solution of Eq. (1) for large values of D and the theoretical results. The agreement is very good both for white and colored noise.

In spite of the good agreement found between our results (9)-(11) and those of the experimental and numerical simulation we should remark here that the



FIG. 1. Variance of the passage time as a function of the pump noise intensity D and correlation time τ $(a_0 = 2.16 \times 10^6 \text{ s}^{-1})$. The full curves refer to the theory [Eq. (11)] and the circles to numerical solutions of Eq. (1).

generating function that we have calculated is for the passage-time distribution and not for the first passage times. Multiple crossing of a given reference value by the process I(t) is a phenomenon which is enhanced by multiplicative noise. The reference values considered here correspond to the time scale in which the system leaves the unstable state. For these values possible multiple crossings occur for times which are close to each other so that the passage-time distribution can be identified for practical purposes with the first-passage-time distribution. For larger reference values the system enters the nonlinear regime dominated by multiplicative noise and multiple crossing becomes important. A characterization of this nonlinear regime is better given in terms of the statistical properties of the intensity. This point of view corresponds to the alternative approach to the study of transient statistics mentioned before. We have calculated the mean value of I and its variance $\delta(t) = \langle I^2 \rangle - \langle I \rangle^2$ from our approximation (4) for the process I(t). A comparison between our theory and a direct simulation of process (1) for the experimental values of the parameters used before is shown in Fig. 2 where very good agreement is found. The effect of the different noise parameters in the anomalous fluctuation phenomenon characterized by the peak of $\delta(t)$ is shown in Fig. 3. A novel feature to be noted is the drastic decrease of fluctuations implied by a colored noise with respect to the white-noise limit $\tau \rightarrow 0$. The value Δ of the anomalous fluctuation is defined as the difference in the value of $\delta(t)$ at its maximum and the asymptotic value of δ as $t \to \infty$: $\Delta = \delta(t_{\max}) - \delta(\infty)$. It is seen in Figs. 2 and 3 that the value of Δ is slightly affected by pump noise. Pump noise must be com-



FIG. 2. The variance of the intensity δ as a function of $a_0 t$ for the experimental values of Ref. 2 $\epsilon = 0.0043$ s⁻¹, $D = 3 \times 10^4$ s⁻¹, $\tau^{-1} = 2.4 \times 10^6$ s⁻¹. The curves refer to the quasideterministic theory and the numerical solution of Eq. (1).



FIG. 3. The calculated variance of the intensity δ as a function of $a_0 t$ and $a_0 \tau$ for $\epsilon = 0.0043 \text{ s}^{-1}$, $D = 3 \times 10^5 \text{ s}^{-1}$.

pared with the average pump parameter. When pump noise is small, as for the experimental values of the parameters in Fig. 2, Δ is slightly reduced with respect to the case in which only quantum noise is present. The effect of larger pump noise is seen in Fig. 3. In this case Δ is strongly reduced for white noise so that anomalous fluctuations disappear for large pump noise with $\Delta \rightarrow 0$. However, this reduction of Δ is compensated by the colored character of pump noise which partially restores the value of Δ . These results and the ones obtained for the variance of the passage-time distribution suggest that experiments on the transient behavior of a dye laser in a region closer to threshold (small a) would be useful to emphasize the effect of pump noise and also to evidence the role played by its finite correlation time.

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 10 We remark that (5) defines a process different from the linear version of the original process (1). For a discussion of this point see Ref. 3.

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